Advanced Online Reprocessing Simulation for Thorium-Fueled Molten Salt Breeder Reactor

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ILLINOIS

Background

Motivation Objectives







2 Methodology

3 Results and discussion



Background

Motivation

Potential Generation IV reactor systems





LEAD FAST REACTOR

GAS FAST REACTOR







VERY HIGH TEMPERATURE REACTOR SUPERCRITICAL WATER REACTOR MOLTEN SALT REACTOR

Motivation Objectives

Why Molten Salt Reactors?



Main advantages of liquid-fueled Molten Salt Reactors (MSRs) [2]

- **1** High average coolant temperature (600-750°C) \Rightarrow high thermal efficiency, hydrogen production, cheap heat energy for chemical industry.
- Ø May operate with epithermal or fast neutron spectrums.
- O Various fuels can be used ($^{235}U,~^{233}U,$ Thorium, U/Pu).
- Liquid fuel has strong negative temperature feedback.
- 6 Liquid fuel drains into tanks in emergency.
- **(6)** High fuel utilization \Rightarrow less nuclear waste generated.
- Online reprocessing and refueling.

Main advantages of Molten Salt Breeder Reactor (MSBR) [3]

- Breed fissile ²³³U from ²³²Th (breeding ratio 1.06).
- 233U, 235U, or 239Pu for the initial fissile loading.
- 3 Thorium cycle limits plutonium and minor actinides.
- Ould transmute Light Water Reactor (LWR) spent fuel.

Motivation Objectives

Molten Salt Reactor Experiment vs Molten Salt Breeder Reactor



Molten Salt Reactor Experiment (MSRE)

- 1 8 MW_{th}
- 2 Fuel salt
 - ⁷LiF-BeF₂-ZrF₄-UF₄
 - ⁷LiF-BeF₂-ZrF₄-UF₄-PuF₃
- $\ensuremath{\mathfrak{S}}$ First use of $^{233}\ensuremath{\mathsf{U}}$ and mixed $\ensuremath{\mathsf{U}}/\ensuremath{\mathsf{Pu}}$
- 4 Single region core
- Operated: 1965-1969 at ORNL



- 1 2.25GW_{th} , 1GW_e
- 2 Fuel salt
 - ⁷LiF-BeF₂-ThF₄-²³³UF₄
 - ⁷LiF-BeF₂-ThF₄-²³³UF₄-²³⁹PuF₃
- Breeding ratio 1.06
- Single fluid/two-region core design
- 6 Chemical salt processing plant







Motivation Objectives

Research objectives



Goals of current study

- Create high-fidelity full-core 3-D model of MSBR without any approximations using the continuous-energy SERPENT 2 Monte Carlo physics software [4].
- Develop online reprocessing simulation code, SaltProc, which expands the capability of SERPENT for simulation liquid-fueled MSR operation [5].
- e) Analyse MSBR neutronics and fuel cycle to find the equilibrium core composition and core depletion.
- ② Compare predicted operational and safety parameters of the MSBR at both the initial and equilibrium states.

Outline



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Input data



Table 1: Summary of principal data for MSBR [SALT TO PUMP (4 TOTAL) Thermal capacity of reactor 2250 MW(t) Net electrical output 1000 MW(e) Net thermal efficiency 44.4% ZONE II-B SLATS ETING BOI Salt volume fraction in central core zone 0.132 ZONE I CHANNEL Salt volume fraction in outer core zone 0.37 GRAPHITE CONTROL RODS REFLECTO 8.2 m^{3} Fuel-salt inventory (Zone I) 10.8 m³ Fuel-salt inventory (Zone II) 3.8 m³ Fuel-salt inventory (annulus) MODERATOR REACTOR Fuel salt components LiF-BeF2-ThF4-233UF4 71.75-16-12-ZONE II-A CHANNEL Fuel salt composition ZONE II 0.25 mole%

Figure 2: Plan view of MSBR vessel [3].

Moderator element geometry (Zone I)



Figure 3: Molten Salt Breeder Reactor Zone I unit cell geometry from the reference [3] (left) and SERPENT 2 (right).





Full-core SERPENT model of MSBR





Figure 4: Plan (left) and elevation (right) view of MSBR model.

Core Zone II





Figure 5: Detailed plan view of graphite reflector and moderator elements.

Online reprocessing method





Figure 6: Flow chart for the SaltProc.

SaltProc capabilities

- Remove specific isotopes from the core with specific parameters (reprocessing interval, mass rate, removal efficiency)
- · Add specific isotopes into the core
- Maintain constant number density of specific isotope in the core
- Time-dependent material feed and removal rates
- Store stream vectors in an HDF5 database for further analysis or plots
- Generic geometry: an infinite medium, a unit cell, a multi-zone simplified assembly, or a full-core

Online reprocessing method





Figure 7: Protactinium isolation with uranium removal by fluorination [3].

Online reprocessing approach

- Continuously removes all poisons, noble metals, and gases.
- ²³³Pa is continuously removed from the fuel salt into a decay tank.

$$^{232}_{90} \text{Th} + ^{1}_{0} \text{n} \rightarrow ^{233}_{90} \text{Th} \xrightarrow{\beta^{-}}_{22.3 \text{ min}} \stackrel{233}{\underset{91}{\rightarrow}} \text{Pa} \xrightarrow{\beta^{-}}_{26.967 \text{ d}} \stackrel{233}{\underset{92}{\rightarrow}} \text{U}$$

The effective cycle times for protactinium and fission products removal [3]

Processing group	Nuclides	Cycle time		
Rare earths	50 days			
	Eu	500 days		
Noble metals	Se, Nb, Mo, Tc, Ru, Rh, Pd, Ag, Sb, Te	20 sec		
Seminoble metals	Zr, Cd, In, Sn	200 days		
Gases	Kr, Xe	20 sec		
Volatile fluorides	Br, I	60 days		
Discard	Rb, Sr, Cs, Ba	3435 days		
Protactinium	²³³ Pa	3 days		
Higher nuclides	²³⁷ Np, ²⁴² Pu	16 years		

Feeds

- ²³²Th (maintained constant)
- ²³³U returned from Pa decay tank (the feed rate assumed equal to ²³³Pa removal rate)

Removals

- ²³³Pa separated into a decay tank
- 100% of other poisons removed

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Effective multiplication factor for full-core MSBR model



Figure 8: k_{eff} during a 20 years depletion simulation.

Fuel salt composition evolution





Figure 9: Normalized number density of major isotopes in the core during 20 years of operation.

Neutron spectrum





- MSBR has a epithermal spectrum which is perfect for thorium fuel cycle
- Spectrum becomes harder due to Pu isotopes accumulation in the core

Figure 10: Neutron spectrum for startup and equilibrium composition (normalized per lethargy)

Power and breeding distribution





Figure 12: $^{232}\mbox{Th}$ neutron capture reaction rate normalized by total flux



Temperature coefficients and control rod worth

Table 2:	Temperature	coefficients	of	reactivity	for	initial	and	equilibrium	state

Reactivity coeffi- cient [pcm/K]	Initial	Equilibrium	Reference [3]		
Fuel salt	-3.22 ± 0.044	-1.53 ± 0.046	-3.22		
Moderator	$+1.61\pm0.044$	$+0.97\pm0.046$	+2.35		
Total	-3.1 ± 0.04	-0.97 ± 0.046	-0.87		

Table 3: Control system rod worth for initial and equilibrium fuel composition

Reactivity parameter	Initial	Equilibrium		
Control (graphite) rod integral worth (cents)	28.215 ± 0.825	28.991 ± 0.773		
Safety (B_4C) rod integral worth (cents)	251.805 ± 0.825	210.992 ± 0.774		
Total reactivity control system worth (cents)	505.762 ± 0.720	424.882 ± 0.805		

²³²Th refill rate





- Fluctuation with various interval and amplitude due to batch-wise removal of strong absorbers
- Feed rate increases during the first 500 days of operation and than steadily reduces due to spectrum hardening and accumulation of absorbers in the core
- Average ²³²Th refill rate throughout 20 years of operation is approximately 2.39 kg/day or 100 g/GWh_e

Figure 13: $^{232}\mathrm{Th}$ feed rate over 20 years of MSBR operation

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Conclusions



This study outcomes

- Full-core fidelity model instead of simplified single-cell model [6] was implemented to precisely describe the two-region MSBR concept design sufficiently to accurately represent breeding in the "blanket"
- Effective multiplication factor slowly decreases from 1.075 and reaches 1.02 at equilibrium after approximately 6 years of operation
- Wide diversity of nuclides, including fissile isotopes (e.g. 233 U, 239 Pu) and non-fissile strong absorbers (e.g. 234 U) keep accumulating in the core
- The neutron energy spectrum is harder for the equilibrium state because a significant amount of fission products were accumulated in the MSBR core
- The total temperature coefficient and reactivity control system efficiency decreases throughout reactor operation
- Average ²³²Th refill rate throughout 20 years of operation is approximately 2.39 kg/day or 100 g/GWh_e which is a good agreement with online reprocessing analysis by Oak Ridge National Laboratory (ORNL)

Future research



Future research effort

- Reprocessing parameters (e.g. time step, feeding rate, protactinium removal rate) optimization to achieve maximum fuel utilization, breeding ratio or safety characteristics
- Verify SaltProc against SERPENT 2 extended for trully continuous online fuel reprocessing simulation
- Overlap a multi-physics model of the MSBR in the coupled neutronics/ thermal-hydraulics code, Moltres [7]

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Processing options for MSR fuels



HUDDOGON H Li Social Na	 Elements that escape from fuel salt Elements that can be removed without processing Elements that can be removed only by chemical processing of fuel salt Max Max<											HEJUM He NEON Ne ARCON ARCON					
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	STRONTION	Y	Zr	Note	Мостностное	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те		Хе
CAESIJJH CS	Ва		Hf	Та	TENSOR	Re	Oswitzen Os	lr Ir	PLATINUM	Au	Hg	TI	Pb	Bi	Ро		Rn
Fr	Ra		Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fi	Uup	Lv	Uus	Uuo
		La	Се	Pr	Nd	Pm	Sm	Eu	Gd	Тр	Dy	Но	Er	Tm	Y	Lu	
		Астини	Th	Ра		NPTUMUM	Ри	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	



BUBBLE GENERATOR AND GAS SEPARATOR for MSBR



Chemical processing facility for MSBR





Multiplication factor dynamics during Rb, Sr, Cs, Ba removal (3435days)



MSBR neutron energy spectrum for different regions





Fissile isotopes producing in MSBR core





MSBR plain view





Generation IV Reactors

I

Goals for Generation IV Nuclear Energy Systems [1]

- Sustainability
- 2 Economics
- 3 Safety and Reliability

Proliferation Resistance and Physical Protection Generation 1

